

FUNDAMENTAL BEHAVIOR OF HUMIC ACID AND KAOLIN IN DIRECT SAND FILTRATION OF SIMULATED NATURAL SURFACE WATER

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ABSTRACT

This article discusses the fundamental behavior of kaolin, humic acid, and aluminum in the primary coagulant PAC, which were included together or separately in three simulated natural surface waters, in direct sand filtration. According to the bench scale experiment, the treatment of water containing humic acid with or without kaolin was found to be much more complex than that of turbid waters. With the filtration of water containing both the materials, at most 31 % of humic acid was removed at a pH level of 7, and 78 % at a pH level of 5 corresponding to a suitable coagulation pH level, with the additions of 25 mg/l of PAC and 0.15 mg/l of an organic polymer. A strong correlation was also noted to exist between the behaviors of humic acid and aluminum.

KEYWORDS

Direct sand filtration; Simulated natural surface water; Humic acid; Kaolin; Poly aluminum chloride (PAC); Aluminum; Polymer; Jar test; Head loss.

INTRODUCTION

Filtration has been one of the most important treatment processes in the production of drinking water for over 100 years. Direct sand filtration without a settling tank was considered as a viable alternative to the conventional sedimentation-filtration treatment systems (Hutchison 1976), and has been usually applied to water sources with low to moderate turbidity levels (Collins *et al* 1987; Wagner and Hudson 1982). This is because all the suspended solids included in influent waters must be stored inside small pore spaces of the filter.

Many surface water sources in Hokkaido, Japan, are of low turbidity, and also colored due to the presence of humic substances, similar to the situation in northern countries. The treatment of such waters, in particular, the water containing humic substances, is considered to be much more complex than that of high turbidity waters, or low turbidity and low temperature waters. Recently there has been increasing concern (Edzward *et al* 1987) to remove organic as well as turbid matter from natural surface waters because some of the humic substances which remain in treated water are found to change through the chlorination process into a trihalomethane which causes serious problems in water quality and to human health (Narkis and Rebhun 1975; Rook 1976). Hence the humic substances should be thoroughly removed in the water purification process.

This study has used three simulated natural surface waters prepared by the additions of either or both humic acid and kaolin to tap water. The study focused on the examination of the fundamental behaviors in effluent waters and also inside the filter of both these materials and aluminum in a coagulant PAC.

EXPERIMENT

Water and Reagents

In order to prepare the three simulated waters for the experiments, the university tap water, quality features of which are listed in Table 1, and the following reagents were used ; (1) kaolin : manufactured by Kanto Chemical Co., Ltd. (2) humic acid : also produced by Kanto Chemical Co., Ltd. In order to dissolve humic acid in a 0.1 N-NaOH solution under flash-mixing, a full day was spent. (3) PAC (Poly aluminum chloride) : The content of Al_2O_3 was 10.5-11.0 % in weight. (4) Polymer : A non-ionic organic polymer with a molecular weight of $1,200 \times 10^3$. A rapid mix of around 20 hours was required to dissolve this polymer. Prior to tests, the prepared polymer solution was diluted to the concentrations specified in the test, and was added to the tap water by use of a micro-tube pump. The polymer was used as a coagulant aid.

TABLE 1 Quality Features of KIT Tap Water

Items	Concentration	Items	Concentration
water temp. (C)	9.5	total residual	(mg/l) 231
pH	6.6	Cl^-	(mg/l) 16.6
turbidity (mg/l)	< 1	NO_3^- -N and NO_2^- -N	(mg/l) 1.93
conductivity ($\mu S/cm$)	94.1	iron	(mg/l) 0.15
alkalinity (mg/l)	57	manganese	(mg/l) nd
hardness (mg/l)	74.5	surfactants(anionic)	(mg/l) nd

Measurements of Materials in Water Samples

Humic acid in the water samples containing only humic acid, was measured by a spectrophotometer using a wavelength of 390 nm and providing a light path of 1 cm, while the level of kaolin in the sample waters containing only kaolin, was determined by use of a turbidimeter providing a light path of 2 cm. Moreover, with regard to the water samples containing both the materials, a new method based on some chemical and physical measurements (Ebie and Amano, 1990) was applied to determine the individual concentrations during the course of the study. Table 2 indicates the relative errors in samples calculated by means of this method. From the table, the relative errors were found to range up to 7-9 % at most, even when the humic acid concentrations were very low. Furthermore, aluminum was also measured by a spectrophotometer, the so called, the "aluminon method" being used.

TABLE 2 Relative Errors in the Measurement of Water Samples Containing Both Humic Acid and Kaolin

Sample mg/l		Measured value mg/l		Relative error %	
kaolin	humic acid	kaolin	humic acid	kaolin	humic acid
20.0	5.00	19.3	4.85	3.5	3.0
20.0	3.00	19.5	3.00	2.5	0.0
20.0	1.00	19.8	1.09	1.0	9.0
2.0	5.00	2.0	4.83	0.0	3.4
2.0	3.00	1.9	3.00	5.0	0.0
2.0	1.00	2.0	1.07	0.0	7.0

Apparatus and Method of Experiment

The apparatus used for filtration experiments is shown in Fig. 1. This employs a direct sand filtration scheme of treatment and three single media filter arrangements. Each filter had a sectional area of 50 cm^2 , the inside of which was packed with silica sand (an effective size of 0.91 mm and a uniformity coefficient of 1.68) with a depth of 60 cm. Piezometer taps allowed incremental head loss measurements to be determined for every

5.0 cm of filter media. In the experiment, at the inlet of the chamber, the pH level of the university tap water was adjusted to 5 or 7, corresponding to the suitable coagulation pH level of either humic acid or kaolin that was chosen on the basis of the jar test results, and then either or both humic acid of 5 mg/l and kaolin of 20 mg/l, and PAC of 7.5 or 25 mg/l as the primary coagulant were also added to the water together with or without the polymer. Small mixers supplied a flash-mix velocity gradient for around 3 minutes. During this period the coagulated water passed under and over the baffles to the outlet section, and then was fed by gravity on to the filters. The filtered water was pumped from below the underdrains at a constant flow of 240 m/d controlled by a needle valve setting throughout the duration of the run. All filter runs were terminated at a head loss of 2.5 m or at a run time of 18 hours.

During filter runs, head loss developments were measured, and small amounts of the influent and effluent water to and from the filter were sampled and analyzed for turbidity and humic acid concentration, the method described earlier being used. In addition, sand samples were also extracted from 14 different depths in the filter after each run, and each amount of turbid matter and humic acid in the deposits captured inside the filter, was determined by the jar test technique which included both floc separation and turbidity measurements. The removal of aluminum was also investigated.

RESULTS AND DISCUSSION

Behavior of Humic Acid and Kaolin in the Jar Test

Prior to the filtration experiments, jar tests were carried out to evaluate effects of the pH of water on the removal of kaolin and humic acid. During the tests, the dosages of humic acid, kaolin and PAC were held at 5, 20 and 25 mg/l, respectively. The tests were initiated under the conditions of a rapid mixing of 100 rpm for 5 min, a slow mixing of 40 rpm for 20 min and settling for 20 min. After the settling step, supernatants of 50 ml were withdrawn from a depth of 5 cm under the water surface in the beaker, and then the pH, turbidity and humic acid of the water samples were measured. The relations between the removal of the materials and the pH established in the tests are illustrated in Fig. 2.

First, in the case of water containing only kaolin, the highest removal rate was 15% at a pH level of 4.5 or lower, but increased greatly to 97-98% at a pH level of 5.5 or higher which was included in the optimal

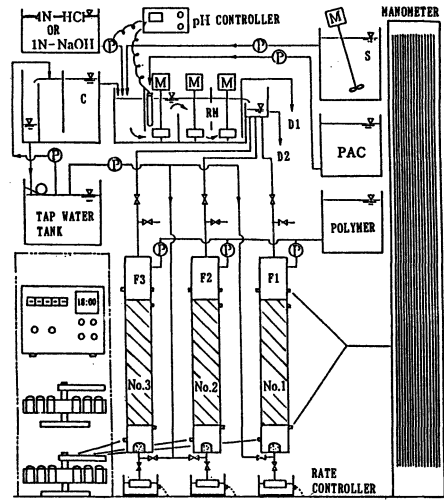


Fig. 1. Schematic Diagram of the Experimental Apparatus

- S : Storage Tank for Humic Acid or Kaolin Suspension
- C : Constant Head Tank
- RM : Rapid Mixing Tank
- D1, D2 : Drain,
- F1, F2, F3 : Filter
- SI1, SI2 : Sampling Instrument of Filter Influent and Effluent

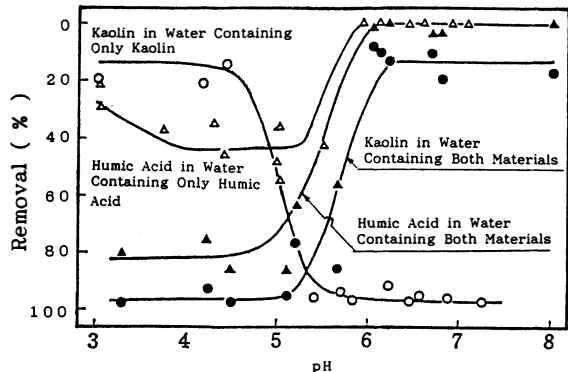


Fig. 2. Relation between Removal of Each Material and pH of Water in Jar Test (Kaolin 20 mg/l, Humic Acid 5 mg/l and PAC 20 mg/l)

coagulation pH range of kaolin with PAC. By contrast, in the case of water containing only humic acid, the highest removal of 40 % was obtained in the pH range of 4.0 to 5.5. Hence, this pH range was considered to be optimal for the coagulation of humic acid, because many brown humic acid flocs were formed in the range. The addition of more coagulant to the water probably leads to higher removal. At a pH level of 6 or higher, no suspended particles were observed in the beaker, and the removal of the humic acid was almost zero under this condition.

On the other hand, in the tests of water containing both the materials, it was noted that the relation of kaolin removal to the pH level was very similar to that of the humic acid. That is, on the acid side of a pH level of 3 to 5, the removal of humic acid increased greatly to 80 % (about 40 % higher than that in water containing only humic acid), and the removal of kaolin as well, to 97 % (over 80 % higher than that in water containing only kaolin). On the contrary, the removal rate of kaolin at a pH level of 6 or higher indicated a significantly low value of 15% (over 80 % lower than that in water containing only kaolin). It was also noted that there was no change in the removal rate of humic acid in the pH range.

These results indicated that on the acid side of pH, kaolin particles were captured inside the mass of humic acid flocs, and even if a pH level of 6 or higher was originally in the coagulation range of kaolin, the kaolin particles could not be coagulated effectively because a much greater amount of PAC would be adsorbed in the humic acid particles than in the kaolin particles.

Head Loss Values across the Filter Bed

Table 3 shows the total head loss values obtained at the end of each filter run. As past studies indicated, the head loss values were greatly affected by filtration variables such as pH levels, type and dosage of coagulant and properties of materials added to waters. At first, when the coagulant dosage was fixed at a PAC dosage of 25 mg/l without the polymer, the filtration of water containing only kaolin, at a pH level of 7, exhibited the considerably higher head loss value of 143.3 cm than the value of 11.4 cm at the pH level of 5, while in the filtration of water containing only humic acid, the head loss value at the pH level of 5 was about 30 % higher than that at a pH level of 7. In addition, with the filtration of water containing both the materials, the higher head loss values occurred at the pH level of 5, not at 7. These head loss values at a

TABLE 3 Conditions and Results of Filtration Experiments

Exp. No	Material & Dosage mg/l	P A C mg/l	Polymer mg	pH	Filtration time hr	Head loss cm	Average removal rate (%)		
							Kaolin	Humic acid	Aluminum
1		7.5	0.00	7	18	56.3	87.1	-	68.8
2	kaolin	7.5	0.05	7	3.5	207.2	98.1	-	78.2
3	20	25.0	0.00	7	18	143.3	52.3	-	61.3
4		25.0	0.00	5	18	11.4	81.6	-	13.7
5		25.0	0.00	7	18	61.6	-	38.9	32.6
6		25.0	0.05	7	18	103.1	-	46.0	42.7
7	humic	25.0	0.15	7	18	130.6	-	51.7	48.7
8	acid	25.0	0.00	5	18	82.3	-	45.5	33.7
9	5	25.0	0.05	5	18	124.1	-	50.8	52.3
10		25.0	0.15	5	5	223.6	-	98.4	86.8
11	kaolin	25.0	0.00	7	18	26.0	69.3	18.5	20.7
12	20	25.0	0.05	7	18	47.3	70.9	21.9	24.0
13	+	25.0	0.15	7	18	111.3	85.4	30.7	30.5
14	humic.	25.0	0.00	5	18	75.1	62.4	51.8	58.1
15	acid	25.0	0.05	5	18	96.4	58.0	60.4	64.2
16	5	25.0	0.15	5	13	204.9	76.1	78.0	75.0

pH level of 5, however, were lower than those in the filtration of water containing humic acid only.

From these results, it can be noted that the production of head loss was strongly related to the optimal coagulation pH level of each material, and that the formation of the humic acid flocs did affect not only the head loss production but also the treated water quality. Moreover, the addition of the polymer to the waters resulted in a substantial increase of head loss developments in all the experiments, and at times led to markedly shorter filtration periods. The use of the polymer therefore should be limited to cases of coarser sand or a multi-medium filter bed, or both, as suggested by previous work.

Behavior of Kaolin, Humic Acid and Aluminum in Filtration of Water Containing Either Humic Acid or Kaolin

Figs. 3 and 4 show the profiles of effluent turbidity and aluminum over time in the filtration of water containing only kaolin at the pH levels of 5 and 7. The removal rates of kaolin at a pH level of 7 were over 95 % for 1.5 or 6 hours from the start of each filter run. The removal rate then decreased rapidly with time, which meant the occurrence of the terminal breakthrough of kaolin. The tendencies of aluminum removal in these experiments were similar to those of kaolin, as shown in Table 3. Hence, at this pH level it is necessary to apply the minimum dosage of PAC to improve removals of both the kaolin and aluminum. On the other hand, the removal rate of kaolin at a pH level of 5 was 50 % at the start of a filter run. However, it increased gradually with time, leading to the high removal rate of 88 % at the end of the filter run, though kaolin particles were in the stable pH range on the jar test basis. In this case, the average of aluminum removal was only 13.7 %. These facts have suggested that in order to enhance aluminum removal, the adjustment of the pH of water to the optimum coagulation range is most important, and even if the pH level of water is not in this range, a proportion of the PAC can make kaolin flocs big enough to be retained in the filter bed.

Figs. 5 and 6 illustrate the relationships of humic acid and aluminum in effluents over time in the filtration of water containing only humic acid. Firstly, the removal of humic acid at a pH level of 7 was around 10 % at the start of the filter run. Thereafter, as the filtration time proceeded, the removal increased to 40-60 % at the filtration time of 6-9 hours. In contrast, the removal of humic acid at a pH level of 5 was as high as around 80 % at the start, and decreased to 15-25 % at the end of the filter run. Because the pH level of 5 was in the optimal coagulation range of humic acid, an excessive number of flocs were reformed, resulting in the sudden occurrence of the break-through of both the humic acid and aluminum. The average removal rates of humic acid, as indicated in Table 3, were slightly higher than those of aluminum at both the pH levels in these

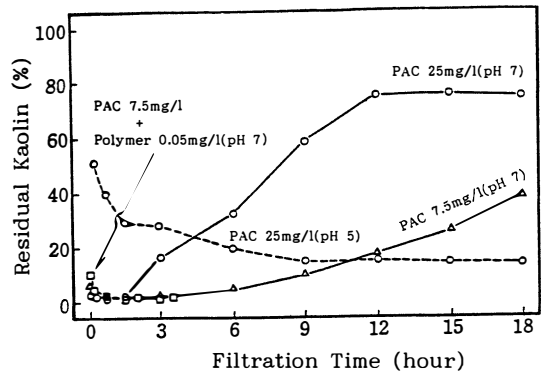


Fig. 3. Behavior of Residual Kaolin (%) in Filtration of Water Containing Only Kaolin

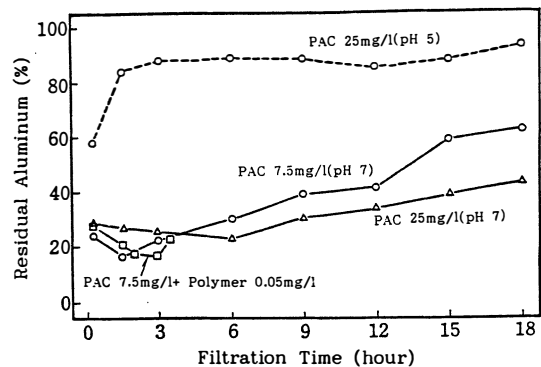


Fig. 4. Behavior of Residual Aluminum (%) in Filtration of Water Containing Only Kaolin

experiments. However, the behavior of both the materials over time were very similar to each other. It was also noted that the removal of humic acid at the initial stage of filtration was almost the same as that in the jar tests. With respect to the effect of the polymer, all the experiments that used the polymer have exhibited considerable effectiveness, namely, the additions of the polymer dosage of 0.15 mg/l together with PAC caused the removal of humic acid, 51.7 %, at a pH level of 7, which was around 13 % higher than in the case of PAC, and at a pH level of 5, a rapid increase of head loss and the corresponding short filter run occurred.

Behavior of Humic Acid and Kaolin in Filtration of Water Containing Both Materials

Figs. 7 to 9 show the results of the filtration of water containing both humic acid and kaolin.

The behaviors of these materials at a pH level of 7 split completely into two parts: the higher removal of kaolin and the lower removal of humic acid. The difference between these removals was marked, namely, 2.6-4.7 fold in the average removal rates that were listed in Table 4. In addition, their tendencies were similar not to jar test results on the water containing both the materials, but to those on the waters containing either. With a PAC dosage of 25 mg/l, the average rate of kaolin removal increased to 85.4 % from 69.3 % by the addition of the polymer of 0.15 mg/l, whereas the average rate of humic acid removal increased to 30.7 % from 18.5 %. From these results, the polymer was found to be more effective than the PAC in increasing the removal rates of both the materials. The behavior of aluminum removal during filter runs at this pH level was very similar to that of humic acid, just as in the cases in the filtration of water containing only humic acid.

On the other hand, at a pH level of 5, both the humic acid and kaolin were found to exhibit relatively high removal rates, in particular in the initial stage of the filter run. However, the head loss production was lower than that in the filtration of water containing only humic acid. This fact indicates that the flocs of the higher density were probably due to the inclusion of kaolin particles.

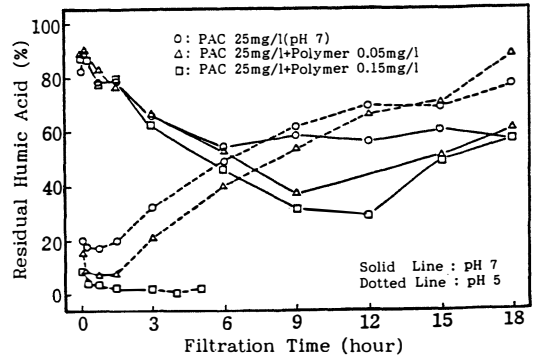


Fig. 5. Behavior of Residual Humic Acid (%) in Filtration of Water Containing Only Humic Acid

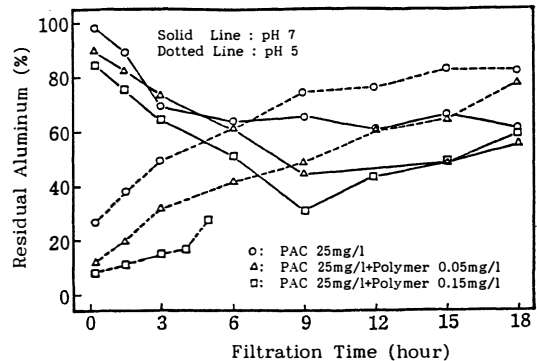


Fig. 6. Behavior of Residual Aluminum (%) in Filtration of Water Containing Only Humic Acid

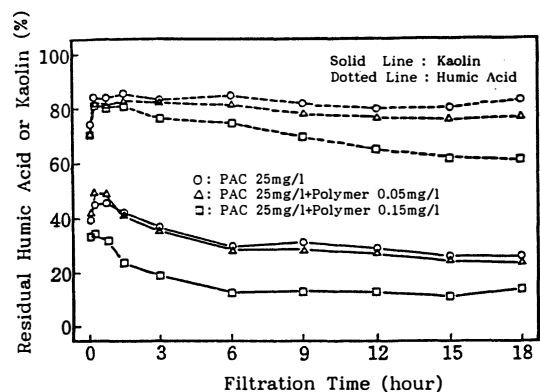


Fig. 7. Behavior of Residual Humic Acid or Kaolin (%) in Filtration of Water Containing Both Materials (pH 7)

Also, the behavior of the removal rate of kaolin, humic acid and aluminum over time was very similar to that of humic acid in the filtration of water containing only humic acid at the same pH level of 5. With respect to effluent concentrations over time, their removal was very high for the initial period of the filtration, but immediately after that, the terminal breakthroughs of all the materials occurred gradually. The effect of the polymer was approximately similar to the results of the other experiments as described earlier. Average removal rates of humic acid were about 10 % higher in the filtration of water containing both the humic acid and kaolin than in the filtration of water containing only humic acid, except in the case with the addition of the polymer of 0.15 mg/l, but slightly lower than the values in the jar tests. If anything, the improvement of the removal rate of the materials can be realized by adopting the optimal coagulant dosage or higher porosity filter bed, or both.

Distribution of Deposits inside the Filter and Ratio of Aluminum to the Deposits

Figs. 10 to 11 demonstrate the distribution of deposits in some experiments, on a weight basis, over the depth of the filter. In the figures, the scale of the axis of abscissa is taken as a common logarithm. Excepting a couple of filtrations where there was too great an increase in head loss, and as a result, markedly short filter runs occurred by the use of the polymer, the distribution of deposits inside the filter are successfully expressed by the following relation.

$$\log_{10}y = -ax + \log_{10}b \quad (1)$$

in which, y denotes the weight of the deposits per unit depth of the filter [mg/cm]; x represents the filter depth [cm]; and "a" and log₁₀b stand for the slope of a line and value of the intercept point with the abscissa respectively.

With respect to "b", the value approximated 10³ mg/l in the cases of kaolin deposits (except the case in which the value of 1x10²-2x10² mg/l was obtained in the filtration of the water containing both the humic acid and the kaolin at a pH level of 5) and 10² mg/l in the cases of the deposits containing humic acid. The adoption of the lower concentration

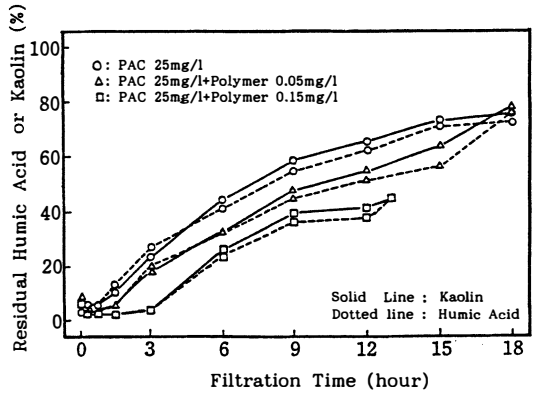


Fig. 8. Behavior of Residual Humic Acid and Kaolin (%) in Filtration of Water Containing Both Materials (pH 5)

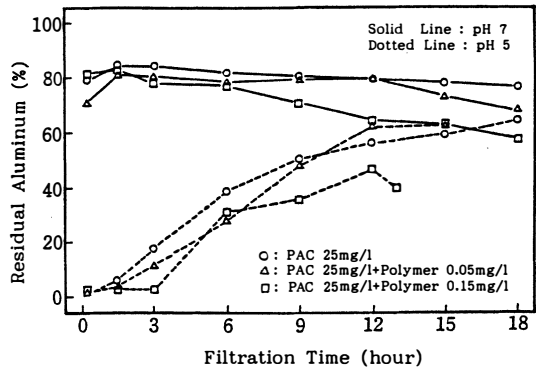


Fig. 9. Behavior of Residual Aluminum (%) in Filtration of Water Containing Both Materials

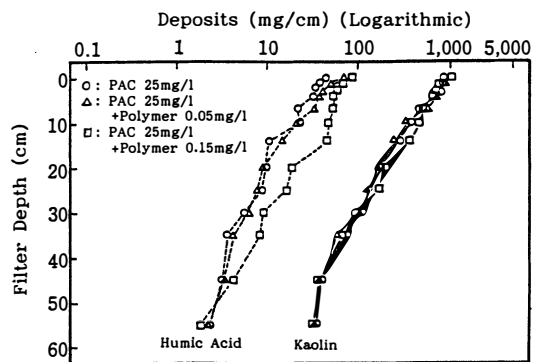


Fig. 10. Distribution of Deposits in Filtration of Water Containing Both Materials (pH 7)

is probably the underlying reason for the humic acid to have smaller "b" values, as seen in these figures. With respect to "a", smaller values occurred at the optimal coagulation pH level of every material in the filtration of water containing either humic acid or kaolin, whereas there was no difference between the "a" values of the materials at both the pH levels of 5 or 7 in the filtration of water containing both the materials. These indicate that the deposits in the filter layer have a particular ratio of humic acid and kaolin.

Table 4 illustrates results on the total deposits and the deposits of the individual material in the filtration of water containing both the materials at the pH levels of 5 and 7. The total deposits at either pH levels of 5 and 7 were approximately equal to each other in these experiments. However, the weight of humic acid contained in the deposit at a pH 5 was 2 to 3 times more than that at pH 7. Hence, average values of the ratio of kaolin and humic acid in the deposits were 4.4 and 14.1 at the pH levels of 5 and 7 respectively. Considering that the aluminum concentration in effluents was high at a pH level of 7, it is suggested that the density of flocs removed in the filter bed at this time was much higher at the pH level of 7 than at the pH level of 5. In addition, the total deposits in the filtration of water containing both the materials showed values markedly larger than those in the filtrations of water containing only humic acid at pH levels of 5 and 7, or only kaolin at a pH level of 7. Accordingly, these facts suggest that in the filtration of water containing both the materials, the pore spaces in the filter can be utilized more effectively than in the filtration of water containing either humic acid or kaolin only.

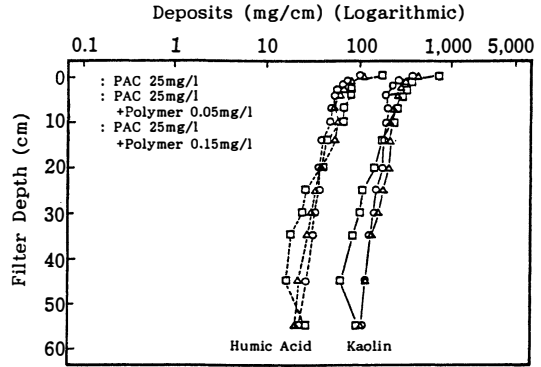


Fig. 11. Distribution of Deposits in Filtration of water Containing Both Materials (pH 5)

TABLE 4 Total Deposits in Filtration of Water Containing Both Kaolin and Humic Acid

Exp. No	Materials added mg/l	pH	Polymer mg/l	Kaolin mg	Humic acid mg	Kaolin-humic acid ratio	Total deposits mg
11		7	0.00	11,977	621	19.29	12,598
12		7	0.05	11,473	718	15.98	12,191
13	kaolin(20)	7	0.15	12,864	1,243	10.35	14,107
14	humic acid	5	0.00	9,100	2,139	4.25	11,239
15	(5)	5	0.05	10,896	2,189	4.98	13,085

* : The filtration time was 13 hours.

Table 5 shows the values of the ratio of aluminum to the total deposits, obtained under various experimental conditions. Unlike previous works, it is noted that the ratios from every filter layer do not change significantly throughout the filter bed. This is probably dependent on the fact that the measurement was made at the filtration time of 18 hours, when considerably high breakthrough had occurred. In the filtration of water containing only kaolin, the ratio is 9 times higher at a pH of 7 corresponding to the suitable coagulation pH level of kaolin, than at the pH level of 5. On the other hand, in the case of filtration water containing only humic acid, values nearly 10 times higher than in the filtration of waters containing both the materials Kaolin only were obtained. This indicates the necessity of a higher coagulant dosage for making humic acid flocs. However, the higher coagulant dosage results in the formation of light and fragile flocs, leading to a low removal rate of humic acid.

TABLE 5 Average Weight Ratio of Aluminum to Turbidity Contained in the Filter Deposit

Materials added (mg/l)	pH	P A C (mg/l)	Polymer (mg/l)	Ratio of Al to turbidity
kaolin (20)	5	25.0	0.00	0.5×10^{-2}
	7	7.5	0.00	1.0×10^{-2}
	7	25.0	0.00	4.5×10^{-2}
humic acid (5)	5	25.0	0.00	11.3×10^{-2}
	5	25.0	0.05	13.7×10^{-2}
	5	25.0	0.15	11.4×10^{-2}
	7	25.0	0.00	18.0×10^{-2}
	7	25.0	0.05	15.6×10^{-2}
	7	25.0	0.15	17.4×10^{-2}
kaolin (20)	5	25.0	0.00	2.2×10^{-2}
+ humic acid(5)	7	25.0	0.00	0.6×10^{-2}

CONCLUSION

The findings from the filtration experiment are summarized as follows.

The production of head loss in the direct sand filtration was highly correlative to the coagulation of materials such as humic acid and kaolin. Hence the filtration of water containing only kaolin exhibited higher head loss values at a pH level of 7, and also in the cases of water containing humic acid only, at a pH level of 5. Moreover, with the filtration of water containing both the materials, the head loss values at a pH level of 5 were 2 to 3 times greater than those at a pH level of 7.

The rate of removal in the jar test was comparable with that at the initial stage of filtration, except in the case of the removal of kaolin in the filtration of water at a pH level of 7, containing both materials. This implies that the difference between the rate of removal in the jar test and that in the filtration will increase, as filtration times proceed.

At a pH level of 5, with respect to the removal rate of humic acid, the filtration of water containing both materials indicated a higher average removal rate than in the filtration of water containing humic acid alone. On the other hand, at a pH level of 7, the removal of each in the filtration of water containing both materials was lower than the removal rate of each material in the filtration of water containing either humic acid or kaolin.

In direct sand filtration, the behavior of humic acid was quite similar to that of aluminum. With the filtration of water containing humic acid only, the behavior of aluminum in the filter effluent over time was substantially similar to that of humic acid, at pH levels of 5 and 7. With the filtration of water containing both materials, the aluminum behavior was very similar to that of both humic acid and kaolin, at a pH level of 5 and also that of humic acid as well, at a pH level of 7.

The total amounts of deposits, which were caught inside the filter bed in the filtration of water containing both materials, were nearly equal to those in the filtration of water containing only kaolin, but were much greater than those in the humic acid filtration. With the filtration of water containing both materials, the deposits at a pH level of 5 were nearly equal to those at a pH level of 7. Meanwhile, the kaolin-humic acid ratio of the deposits was 4.4 at a pH level of 5, and 14.1 at a pH level of 7. These results suggest that the composition of the deposits changed considerably with the pH level of water.

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